

**Written Amendment**  
(Amendment based on Section 11)

To Director-General of the Japanese Patent Office

**1. Identification of the International Application**  
PCT/JP2004/000463

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**4. Object of Amendment: Claims**

**5. Contents of Amendment**

(1) As shown in a separate sheet, we amend Claim 1 on page 17 (translation: page 21) by inserting "the binder comprises an aqueous resin and a rubber-based resin," after "A negative electrode for lithium secondary batteries, comprising a negative active material and a binder, wherein".

(2) As shown in a separate sheet, we amend Claim 6 on page 18 (translation: page 22).

(3) As shown in a separate sheet, we amend Claim 7 on page 18 (translation: page 22) by inserting "comprising an aqueous resin and a rubber-based resin" after "in the presence of a binder"

(4) As shown in a separate sheet, we cancel Claim 12 on page 19 (translation: page 23).

(5) As shown in a separate sheet, we amend Claim 13 on page 19 (translation: pages 23 to 24) by inserting "the binder comprises an aqueous

resin and a rubber-based resin,” after “the negative electrode comprises a negative active material and a binder.”

(6) As shown in a separate sheet, we amend Claim 18 on page 20 (translation: page 25).

(7) As shown in a separate sheet, we add Claim 19 on page 20 (translation: page 25).

#### 6. List of appended documents

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|---|----------|
| (1) New page 17 (translation: pages 21 to 22) and<br>New page 17/1 (translation: page 22), Claims | one each |
| (2) New page 18 (translation: pages 22 to 23), Claims   | one      |
| (3) New page 19 (translation: pages 23 to 25), Claims   | one      |
| (4) New page 20 (translation: pages 24 to 25), Claims   | one      |

## CLAIMS

1. (Amended) A negative electrode for lithium secondary batteries,  
comprising a negative active material and a binder,  
5        wherein the binder comprises an aqueous resin and a rubber-based  
resin,  
the negative active material comprises graphite A and graphite B,  
shapes of primary particles of the graphite A are spherical or elliptical,  
an average particle diameter of the primary particles of the graphite A  
10    ranges between 10  $\mu\text{m}$  and 30  $\mu\text{m}$  inclusive,  
sizes of crystallites of the graphite A in a direction of a c-axis are smaller  
than 100 nm and tap density of the graphite A is 1.0  $\text{g}/\text{cm}^3$  or higher,  
shapes of primary particles of the graphite B are flat,  
an average particle diameter of the primary particles of the graphite B  
15    ranges between 1  $\mu\text{m}$  and 10  $\mu\text{m}$  inclusive, and  
sizes of crystallites of the graphite B in a direction of a c-axis are 100  
nm or larger.
2.        The negative electrode for lithium secondary batteries according to  
20    Claim 1, wherein at least a part of surfaces of the graphite A is further  
covered with non-graphite carbon.
3.        The negative electrode for lithium secondary batteries according to  
Claim 1,  
25        wherein,  $I_{1350}$  denotes Raman intensity at approximately  $1350\text{cm}^{-1}$ ,  
 $I_{1580}$  denotes Raman intensity at approximately  $1580\text{cm}^{-1}$  and a R-value of  
Raman spectrum is obtained by a formula:  $R=(I_{1350}/I_{1580})$ ,  
a R-value of Raman spectrum of the graphite A is 0.4 or larger when  
the graphite A is excited by an Ar laser with a wavelength of 5145 Å.

4. The negative electrode for lithium secondary batteries according to Claim 1, wherein the primary particles of the graphite B aggregate or bond so as to form secondary particles, and an average particle diameter of the secondary particles ranges between 10  $\mu\text{m}$  and 30  $\mu\text{m}$  inclusive.

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5. The negative electrode for lithium secondary batteries according to Claim 1, wherein a weight proportion of the graphite A ranges between 10 wt% and 90 wt% inclusive, with respect to a sum weight of the graphite A and the graphite B.

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6. (Amended) The negative electrode for lithium secondary batteries according to Claim 1, wherein paint-film density of the negative electrode for lithium secondary batteries is 1.5 g/cm<sup>3</sup> or higher.

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7. (Amended) A method for manufacturing a negative electrode for lithium secondary batteries comprising the steps of:

preparing graphite A of which shapes of primary particles are spherical or elliptical, an average particle diameter of the primary particles ranges between 10  $\mu\text{m}$  and 30  $\mu\text{m}$  inclusive, sizes of crystallites in a direction of a c-axis are smaller than 100 nm, and tap density is 1.0 g/cm<sup>3</sup> or higher;

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preparing graphite B of which shapes of primary particles are flat, an average particle diameter of the primary particles ranges between 1  $\mu\text{m}$  and 10  $\mu\text{m}$  inclusive, and sizes of crystallites in a direction of a c-axis are 100 nm or larger;

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preparing paint by mixing the graphite A and the graphite B in the presence of a binder comprising an aqueous resin and a rubber-based resin, and a solvent; and

applying the paint on a collector, drying the paint and then performing a pressure forming treatment.

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8. The method for manufacturing the negative electrode for lithium secondary batteries according to Claim 7, wherein at least a part of surfaces of the graphite A is further covered with non-graphite carbon.

5 9. The method for manufacturing the negative electrode for lithium secondary batteries according to Claim 7,

wherein,  $I_{1350}$  denotes Raman intensity at approximately  $1350\text{cm}^{-1}$ ,  $I_{1580}$  denotes Raman intensity at approximately  $1580\text{cm}^{-1}$  and a R-value of Raman spectrum is obtained by a formula:  $R=(I_{1350}/I_{1580})$ ,

10 a R-value of Raman spectrum of the graphite A is 0.4 or larger when the graphite A is excited by an Ar laser with a wavelength of  $5145\text{ \AA}$ .

10. The method for manufacturing the negative electrode for lithium secondary batteries according to Claim 7, wherein the primary particles of  
15 the graphite B aggregate or bond so as to form secondary particles, and an average particle diameter of the secondary particles ranges between  $10\text{ }\mu\text{m}$  and  $30\text{ }\mu\text{m}$  inclusive.

11. The method for manufacturing the negative electrode for lithium  
20 secondary batteries according to Claim 7, wherein a weight proportion of the graphite A ranges between 10 wt% and 90 wt% inclusive, with respect to a sum weight of the graphite A and the graphite B.

12. (Cancelled)

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13. (Amended) A lithium secondary battery, comprising a positive electrode, a negative electrode and nonaqueous electrolyte,

wherein the negative electrode comprises a negative active material and a binder,

30 the binder comprises an aqueous resin and a rubber-based resin,

the negative active material comprises graphite A and graphite B,  
shapes of primary particles of the graphite A are spherical or  
elliptical,

an average particle diameter of the primary particles of the graphite  
5 A ranges between 10  $\mu\text{m}$  and 30  $\mu\text{m}$  inclusive,

sizes of crystallites of the graphite A in a direction of a c-axis are  
smaller than 100 nm and tap density of the graphite A is 1.0 g/cm<sup>3</sup> or higher,  
shapes of primary particles of the graphite B are flat,

an average particle diameter of the primary particles of the graphite  
10 B ranges between 1  $\mu\text{m}$  and 10  $\mu\text{m}$  inclusive, and  
sizes of crystallites of the graphite B in a direction of a c-axis are 100  
nm or larger.

14. The lithium secondary battery according to Claim 13, wherein at least  
15 a part of surfaces of the graphite A is further covered with non-graphite  
carbon.

15. The lithium secondary battery according to Claim 13,  
wherein,  $I_{1350}$  denotes Raman intensity at approximately 1350cm<sup>-1</sup>,  
20  $I_{1580}$  denotes Raman intensity at approximately 1580cm<sup>-1</sup> and a R-value of  
Raman spectrum is obtained by a formula:  $R=(I_{1350}/I_{1580})$ ,

a R-value of Raman spectrum of the graphite A is 0.4 or larger when  
the graphite A is excited by an Ar laser with a wavelength of 5145 Å.

25 16. The lithium secondary battery according to Claim 13, wherein the  
primary particles of the graphite B aggregate or bond so as to form secondary  
particles, and an average particle diameter of the secondary particles ranges  
between 10  $\mu\text{m}$  and 30  $\mu\text{m}$  inclusive.

30 17. The lithium secondary battery according to Claim 13, wherein a

weight proportion of the graphite A ranges between 10 wt% and 90 wt% inclusive, with respect to a sum weight of the graphite A and the graphite B.

18. (Amended) The lithium secondary battery according to Claim 13,  
5 wherein paint-film density of the negative electrode is 1.5 g/cm<sup>3</sup> or higher.

19. (Added) The lithium secondary battery according to Claim 13,  
wherein the nonaqueous electrolyte comprises vinylene carbonate.